Supporting Information

From Spin Coating to Zone Casting: Process Dependence and Challenges in Scaling Quasi-2D Perovskite Films

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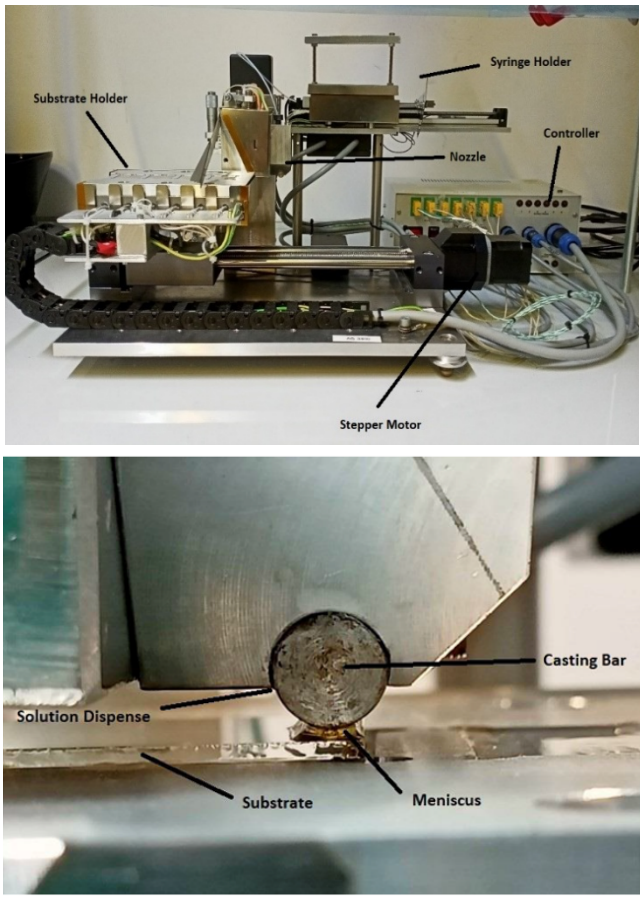

Figure S1. Photographs of the zone-casting instrument (up) and the formation of the meniscus between the cylindrical bar and the substrate (down).

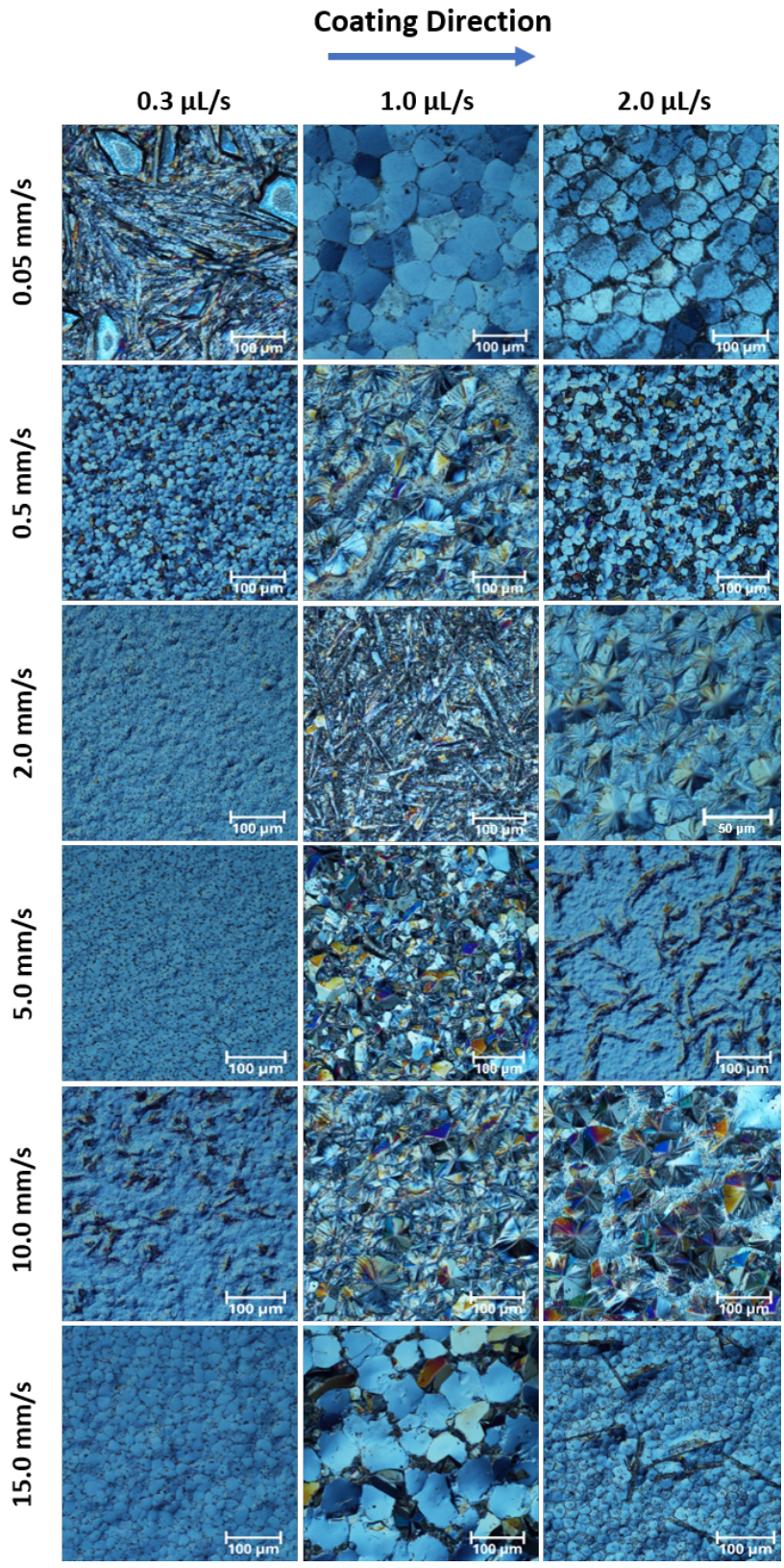

Figure S2. Polarized optical microscopy images of the zone-casted thin films under different injection rates (horizontal axis) and casting speeds (vertical axis). The films were cast over glass substrates. The films were cast using a 9:1 DMF:DMSO solvent mixture and solution-substrate temperature of 60 °C

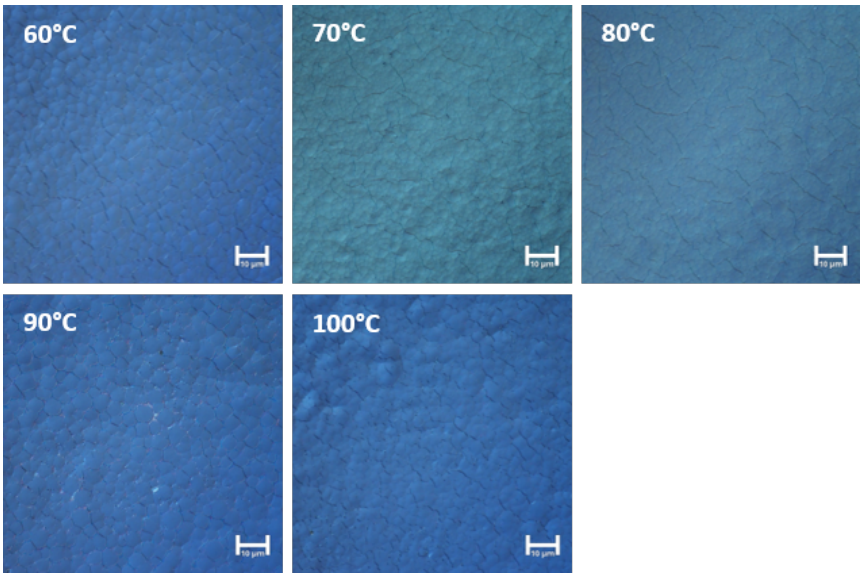

Figure S3. Polarized optical microscopy images of the zone-casted thin film under optimized conditions (nozzle height= 1mm, injection rate= 0.3 µL s^{-1}, casting speed= 5 mm s^{-1}). The films were cast using a 9:1 DMF:DMSO solvent mixture and solution-substrate from 60 to 100° C.

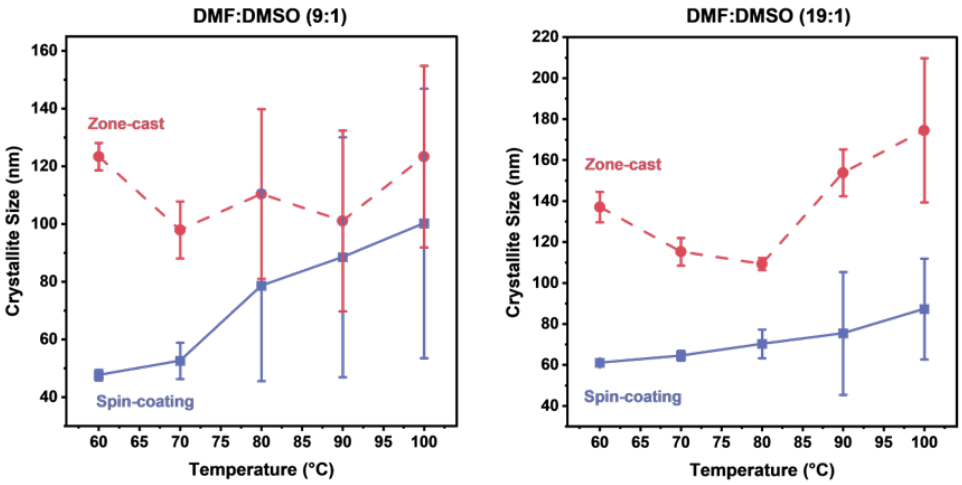

Figure S4. Evolution of the crystallite size estimated from the XRD peak broadening analysis using the FWHM values of the main diffraction reflections for the films processed at 60 – 100° C solution-substrate temperatures and 9:1 and 19:1 DMF:DMSO solvent ratios. The crystallite sizes were calculated from the diffraction peaks using the Scherrer equation, comparing spin-coated (straight lines) and zone-cast (dashed lines) films.

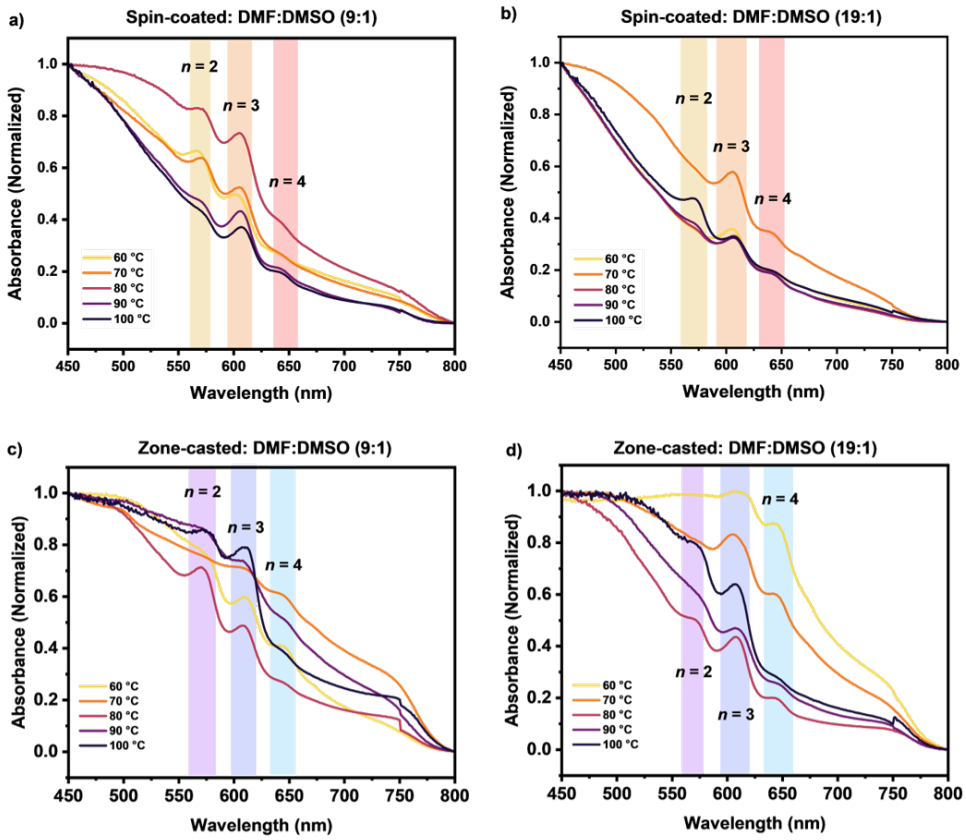

Figure S5. Normalized UV-vis spectra of films fabricated at different solution-substrate temperatures by spin-coating and zone-casting using a,c) 9:1 and b,d) 19:1. DMF:DMSO ratios.

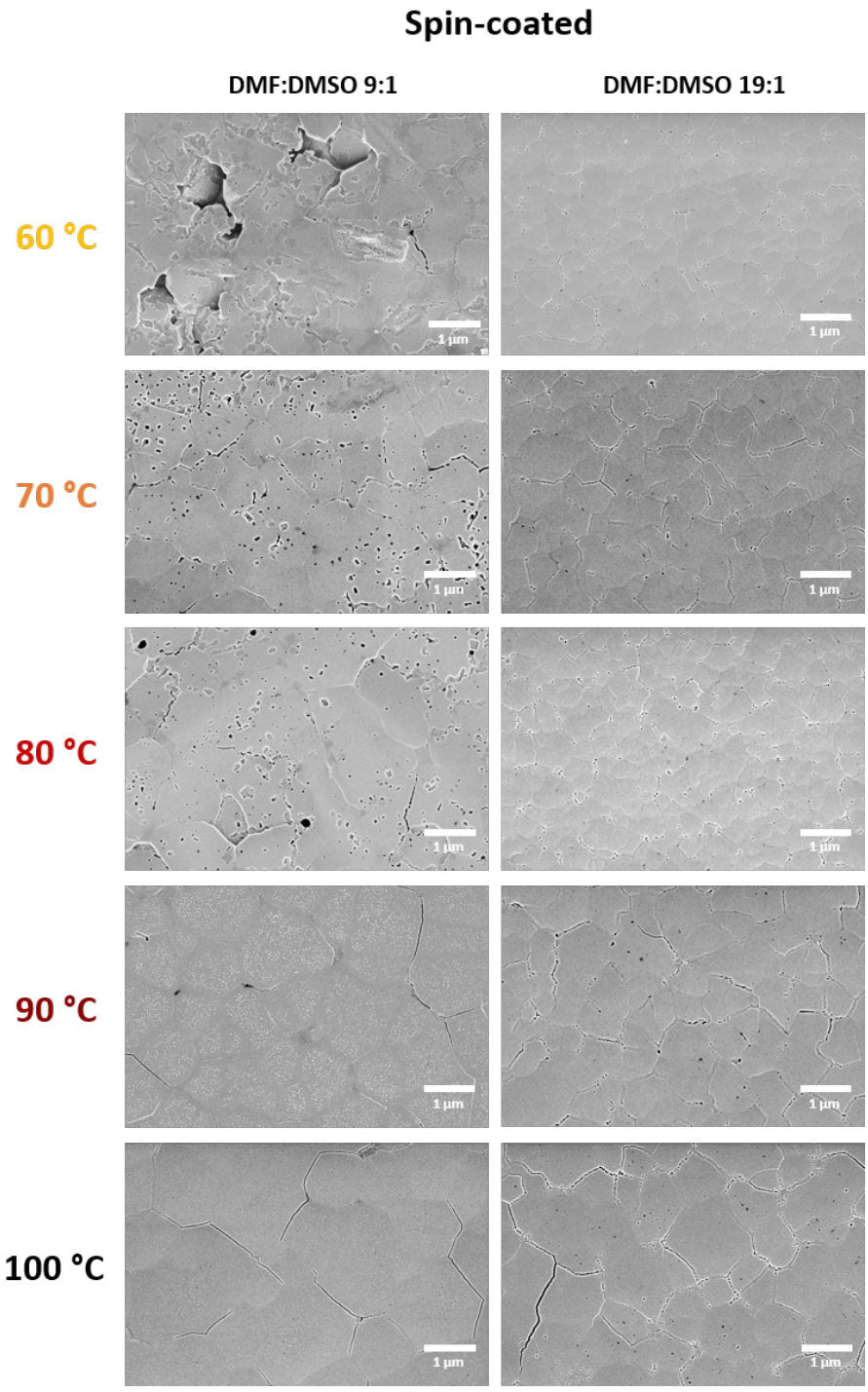

Figure S6. SEM images of thin films prepared by spin-coating from precursor solutions with 9:1 and 19:1 DMF:DMSO volume ratios (Magnification = 50.00K x, detector = InLens) with visible perovskite grains and boundaries.

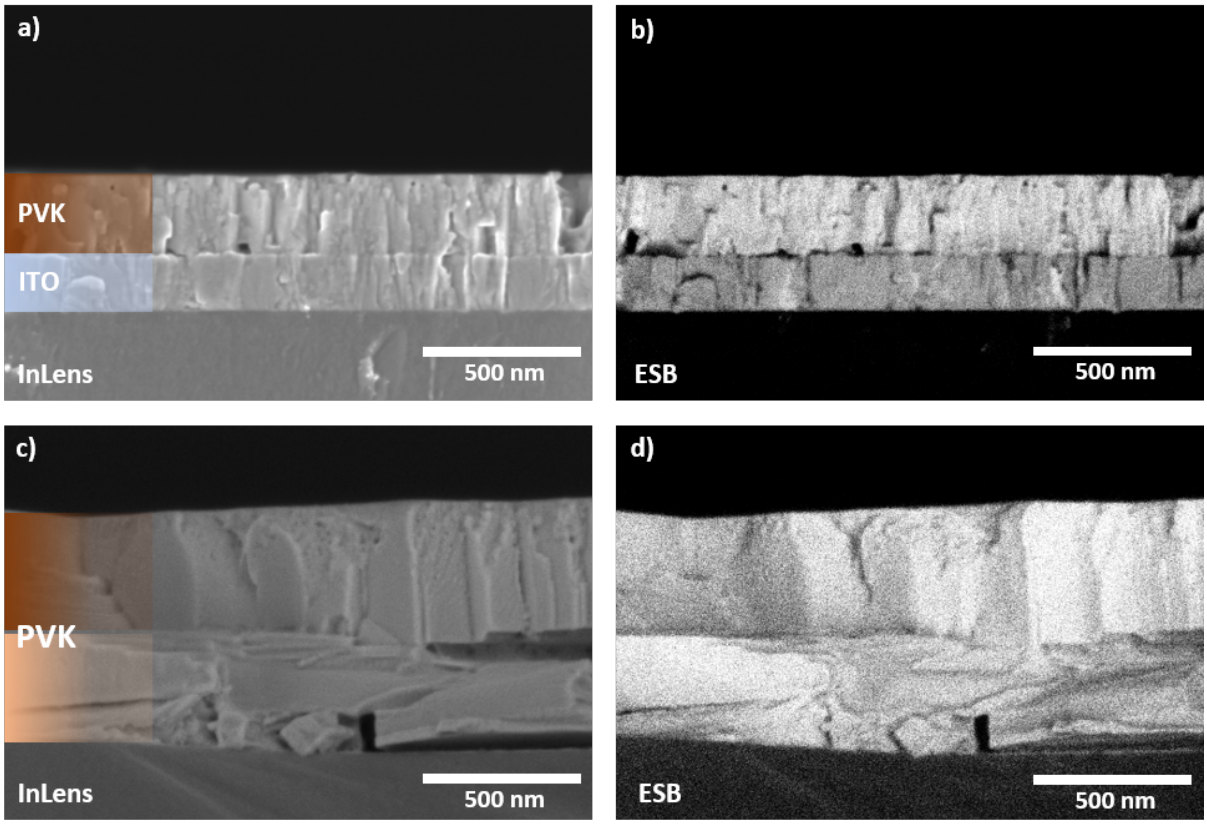

Figure S7. Cross-sectional SEM micrographs of quasi-2D perovskite thin films fabricated by spin-coating (a, b) and zone-casting (c, d), captured under different detectors. The images reveal that the zone-cast films exhibit visible phase segregation. In contrast, spin-coated films display an intermixed phase distribution throughout the layer.

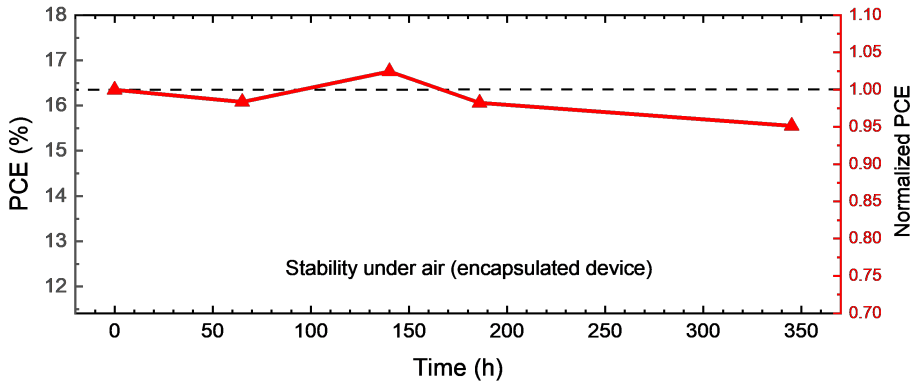

Figure S8. Stability of the champion solar cell fabricated under 19:1 DMSO and solution-substrate temperature of 60°C, and kept under air for over 350 h. The data shows the solar devices remain 95% of its initial performance.